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Publication date:
2013

Document Version
Publisher's PDF, also known as Version of record

NKS-273
ISBN 978-87-7893-346-1

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January 2013

Abstract

Naturally occurring radionuclides (NORs) are the major contributors to the total effective dose of ionizing radiation of the population. Especially hazardous are the decay products of U: ^{210}Pb, ^{210}Po and ^{226}Ra in soils, water and plants. The most important exposure route to ^{210}Pb and ^{210}Po is through wild gathered food. Some studies show that ^{210}Po and also some other NORs accumulate from uranium-rich grounds in mushrooms. In berries the levels are usually lower. In Finland, Sweden and Norway there are sites enriched in NORs. In these areas e.g. the ^{210}Po levels in certain edible mushroom species may be as high as a few hundred Bq/kg, leading to effective doses of several mSv/year among certain consumer groups. The intake of wild forest products varies greatly among the population, but the public should be informed of the exposure risk and ways to minimize it. In this study, NORs and stable metals are analysed in forest soils and in common edible mushrooms and berries. Transfer factors are calculated and dose estimates from consumption of these products made. Based on the measurement data, it is estimated if highly exposed groups exist, and ways of communication with these groups will be discussed. The practical work started during summer-autumn 2012, when soils, berries and mushrooms were sampled at several sites in Finland, Norway, Denmark and Sweden, and pre-treatments carried out in the local laboratories. All sampling and pre-treatment methods were agreed by all partners and ISO standards were used when applicable, to make all stages of the work harmonized between partners. For the analytical work, samples will be sent to partners with suitable analytical facilities. The analyses will be done during the year 2013 and the results reported in the final report and in scientific publications.

Key words

Naturally occurring radionuclide, exposure, effective dose, transfer factor, berry, mushroom

NKS-273
ISBN 978-87-7893-346-1

Electronic report, January 2013
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Interim Report from the NKS-B BERMUDA activity

(Contract: AFT/B(12)8)

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1. Introduction

Naturally occurring radionuclides (NORs) are the major contributors to the total effective dose of ionizing radiation of the world population [1–3]. This natural dose is primarily generated by external and internal exposures from uranium (U) and thorium (Th) and their decay products. Key problems are associated with transport of U and Th and their daughters (especially polonium (Po) and lead (Pb) isotopes) in aquatic and terrestrial ecosystems (Figure 1). Radionuclides are transferred from the site by air emissions, by leaching and run-off water, and from soils into plants, animals and finally to man. Substantial work associated with the evaluation of the radioecological situation in U deposit regions and in vicinity of U mines has been performed in several countries such as Brazil, Australia, Europe, USA and Canada [4, 5]. Special attention is paid to the most hazardous decay products of U; ^{210}Pb and ^{210}Po and ^{226}Ra in soils, waters and plants [6, 7].

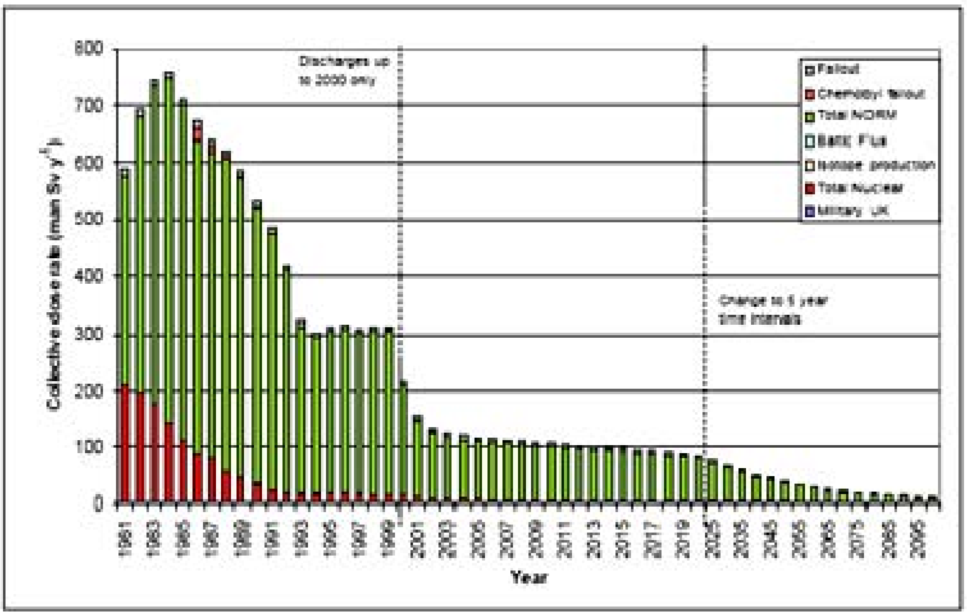

Figure 1. The calculated Collective dose rates by source to the European Union population due to discharges show that the majority of the dose is due to NORMs [1].

NORs that do not originate from industrial discharges or emissions cause also significant doses to the population, most notably radon in indoor air. Also, ground water may act as a significant pathway for exposure and therefore uranium series radionuclides have been extensively studied in the in drinking water of the Nordic Countries [8–10]. Natural radioactivity in foods, however, has been much less investigated and only few studies are available, mainly concentrating on wild gathered food. This is naturally due to the legislation; there are no guideline values or maximum levels regarding NORs in food.

The concentrations of NORs in wild berries have generally been low but surprisingly high concentrations of ^{210}Po have been documented in some mushroom species compared to its grandparent, ^{210}Pb. Vaaramaa *et al.* [11] reported ^{210}Pb and ^{210}Po concentrations in berries and mushrooms at two sites in Finland (Table1). It was obvious that mushrooms can efficiently accumulate ^{210}Po from the soil/litter as the concentrations in were 12–2200 Bq/kg (d.w.) while ^{210}Pb concentrations ranged only 1.5–16 Bq/kg (d.w.).

Table 1. NOR concentrations (in dry weight) in blueberry, lingonberry and various mushrooms at two forest sites [11].

Site	Sample	^{210}Pb	^{210}Po
1	Blueberry Lingonberry Mushrooms	1.7 3.2 1.5–16	2.5 7.5 12–**2200**
2	Blueberry Lingonberry Mushrooms	0.7 1.4 3.3–9.8	3.2 2.2 7.1-**860**

Paukkajavaara uranium mine and mill was commissioned in 1958. After quarrying 31 kt of ore, the mine was shut down in 1961 as unprofitable. The waste rock and tailings were left at the site uncovered. During 1985–1986, blueberry, lingonberry and fungi samples were collected at the mining site, in its vicinity and 12 km south from the site (reference site) [12]. Again, highest ^{210}Po concentrations were in mushrooms (Tables 3–5).

Table 3. NOR concentrations (Bq/kg fw) in blueberry at Paukkajavaara uranium mining site. The dry matter of blueberry is generally 13%.

	At the mine	Vicinity	Reference site
^{238}U			
^{228}Ra	0.61	0–0.35	
^{226}Ra	2.3	0.95–14	0.31–0.40
^{210}Pb	0.20	0.33–0.54	0.40–0.41
^{210}Po	0.30	0.11–0.33	0.25–0.40

Table 4. NOR concentrations (Bq/kg fw) in lingonberry at Paukkajavaara uranium mining site. The dry matter of lingonberry is generally 14%.

	At the mine	Vicinity	Reference site
^{238}U			
^{228}Ra	0.71–2.6	0–0.28	
^{226}Ra	5.5–7.2	0.56–1.1	0.47
^{210}Pb	0.51–1.1	0.42–0.70	0.42
^{210}Po	0.34–0.75	0.28–0.42	0.28

Table 5. NOR concentrations (Bq/kg fw) in boletus at Paukkajavaara uranium mining site. The dry matter of boletus is generally 9%.

	At the mine	Vicinity	Reference site
^{238}U	0.71		
^{228}Ra	0.26	0.43	
^{226}Ra	8.9	0.39	0.52
^{210}Pb	7.4	0.62	0.20
^{210}Po	**460**	7.2	7.8

Phosphate and niobium ores in Sokli contain above average concentrations of NOR's. In phosphate ore, the mean ^{238}U and ^{232}Th concentrations are 310 and 533 Bq/kg, respectively. In the niobium ore, the mean concentrations are 1000 and 4000 Bq/kg, respectively. STUK has carried out a radiological baseline environmental assessment at the site in 2008–2009. Decision about commissioning the mine has not yet been made. The results relating to wild berries and mushrooms are presented on table 6–7.

Table 6. NOR concentrations (Bq/kg dry weight) in wild berries and mushrooms at Sokli, a prospective phosphate/niobium mining site.

	Berries (2008)	Berries (2009)	Mushrooms (2008)	Mushrooms (2009)
^{238}U	<mda	<mda	92	<mda
^{228}Ra	0.6–2.6	1.1–2.7	2.2	<mda
^{226}Ra	0.5–12	0.6–2.0	<mda–58	<mda

Table 7. NOR concentrations (Bq/kg fresh weight) in wild berries and mushrooms at Sokli, a prospective phosphate/niobium mining site. The highest ^{210}Po concentrations were found in boletus (130–830 Bq/kg fw). The dry matter of boletus is generally 9%.

	Berries (2008)	Berries (2009)	Mushrooms (2008)	Mushrooms (2009)
^{210}Pb	1.4–2.5	1.1–2.8	1.9–22	4–19
^{210}Po	1.0–4.9	0.6–1.2	6–**830**	6–236

Talvivaara mine started of nickel and zinc production in 2008. Separation of uranium by bioheap leaching is now being commissioned. STUK is carrying out a radiological baseline environmental assessment at the site in 2010–2012. The results attained so far are on Table 8.

Table 8. NOR concentrations (Bq/kg dry weight) in mushrooms and berries near Talvivaara mine

	Blueberry	Lingonberry	Mushrooms
^{238}U	<mda	<mda	<mda
^{228}Ra	1.7–2.5	1.3–11	<mda
^{226}Ra	1.4–4.5	1.4–5.3	<mda–2.6

In Sweden, Johansson *et al.* [13] studied U and Th in fungi in the Forsmark area. The uranium concentration in fungal fruitbodies was 0.005–0.2 mg/kg d.w. whereas thorium concentration was lower, 0.001–0.013 mg/kg d.w.

Gwynn *et al.* studied activity concentrations of wild berries and mushrooms in northern Norway [14]. The mean activity concentrations of ^{210}Po and ^{210}Pb ranged 0.6–2.6 and 1.2–4.1 Bq/kg (d.w.) for various species of berries. Similarly to findings of Vaaramaa *et al.*, samples of bolete and russule exhibited moderate ^{210}Pb concentrations, in the range of 1–8 Bq/kg (d.w.). Again, much higher ^{210}Po concentrations in bolete, ranging 20–500 Bq/kg (d.w.), were found. In russule, the concentrations ranged 3–9 Bq/kg (d.w.).[1]

Data on NOR concentrations in berries and mushrooms are available from some countries. Davé *et al*. [15] reported ^{226}Ra levels in blueberries ranging 20–290 Bq/kg dry weight in a

[1] At the time of planning the BERMUDA project, these Norwegian data were not availble.

survey near decommissioned Canadian uranium mine. In Brazil, in a region of elevated natural radioactivity, the NOR concentrations (U- and Th-series) in mushrooms levels were 20–60 Bq/kg [16]. Eckl *et al*. [17] could not detect any ^{238}U (MDA 90 Bq/kg d.w.) or ^{226}Ra (MDA 50 Bq/kg dw) in several mushroom samples from Austria. Some samples contained 30–70 Bq/kg d.w. of ^{210}Po while most were below detection limit of 10 Bq/kg. Fungi sampled in three areas in France were analyzed by gamma spectrometry for their concentrations ^{210}Pb and ^{226}Ra [18]. Activity concentrations of ^{210}Pb were in the range <1.76–36.5 Bq/kg d.w. Activity concentrations of ^{226}Ra were consistently lower, often by one order of magnitude. Baeza *et al.* [19–21] and Guillen *et al.* [22] have studied NOR concentrations in mushrooms as well as soil-to-fungi transfer and bioavailable fraction during ingestion in Spain. The ^{210}Pb detected in 30 species of fungi ranged 0.75–202 Bq/kg d.w.

Unlike in Denmark, there are sites enriched in NORs in Finland, Sweden and Norway. The Fen in Norway, for example, contains significant levels of NORs and the outdoor gamma doses are among the highest in Europe. The Fen area contains both high levels of Th, U and daughter nuclides, in addition to enhanced levels of other metals [23–25]. Extensive mining (iron and niobium) have been conducted in these areas in early years. In 2008, the Norwegian Thorium Committee stated that there is a need for radiation protection regulation when it comes to mining and milling in such areas [26]. Before May 2009, natural occurring radionuclide material was not regulated in Norway, but the new Norwegian Pollution Act started to act January 2011. The Act highlights the need for science-based assessment for the Fen area.

The maximum permitted level of ^{137}Cs in mushrooms (600 Bq/kg) leads to 0.1 mSv effective dose from about 10 kg of consumption. If the ^{210}Po levels in mushrooms are high (such as in Boletus, up to a few hundred Bq/kg), the same consumption would lead to effective doses of several millisieverts per year. If we consider the dose constraint of 1 mSv per year for public exposure, it is well possible that among certain consumer groups this is exceeded and the public should be informed about this. As the $^{210}Po/^{210}Pb$ activity ratios have generally been high, up to 300, the high ^{210}Po levels in mushrooms can be reduced by preservation (desiccation, freezing) which would offer an easy method for mitigating the public exposure.

In this study we analyse ^{210}Pb, ^{210}Po, stable metals, uranium, thorium as well as ^{226}Ra and ^{228}Ra in some common edible fungi, blueberries and lingonberries and make dose estimates to the Nordic population from consumption of these forest products. In order to supplement the sparse data on transfer factors, also soil sampling at a few sites are included. Samples will be collected at sites where elevated natural radioactivity is found as well as sites representing average levels.

The specific aims of the project are:

1. to produce data on natural radioisotope levels in wild fungi and berries in Nordic countries
2. based on 1, assess relevant doses to the Nordic population
3. to complement data on soil-to-fungi / soil-to-berry transfer factors
4. to find out if highly exposed groups exists
5. to think of proper ways of risk communication if groups among whom 1mSv/y effective dose is exceeded
6. to be prepared for possible questions about levels in commercially marketed/exported products
7. to share and strengthen know-how in analytics and environmental behaviour of natural radioisotopes among the Nordic Countries

This will be accomplish by:
1. collecting all relevant information concerning natural radioisotopes in wild mushrooms and berries in the Nordic Countries
2. collecting data on consumption rates of these products
3. collecting several representative samples of mushrooms, berries and soils in Finland, Sweden, Norway and Denmark
4. sharing our facilities and analytical capabilities in order to measure the challenging natural radioisotopes in these samples cost-effectively and with high precision

The specific outcomes of the project will be:
1. Interim report in which present data on levels and consumption rates are reported
2. Final report in which the results are presented and relevantly discussed
3. A scientific article about transfer
4. A scientific article about levels and exposure

2. Consumption rates

Intake of wild berries and mushrooms varies greatly among the population. According to a Danish investigation, 56% of the population eat wild berries and only 34% eat wild mushrooms [27]. There were people who consume great amounts of forest produce, as much as 20 kg of wild mushroom per year. According to The National FINDIET 2007 survey, which was conducted by 48-hour dietary recall, only 4 and 7% of men and women consumed foods containing mushrooms [28]. According to three separate surveys, 64–82% of Finnish households use wild mushrooms in their diet [29–31]. Wild berries were used by 94% of Finnish households, on average 8.3 kg per capita annually [29]. According to Saastamoinen 60% of households collected berries themselves [32]. According to Rantakokko [33], the most consumed berries were blueberry and lingonberry.

Sparse existing data on consumption of wild mushrooms regarding the average Norwegian population suggested only 0.24 kg per capita annually [34]. However, numerous $^{137,134}Cs$ monitoring studies in Central Norway demonstrated consumption at level 2.7 kg per capita annually in some population parts. Wild berries are used in Norwegian diet at individual level ranges from 2.6 to 4.5 kg per year. More specific, for selected individuals in Snåsa, Norway, with presumed high consumption of locally produced foods, it was found an annual consumption of wild mushrooms at aprox. 2.7 kg per household. For the same group, the intake of wild berries was annually approx. 24.5 kg per household. In the same study, a population of reindeer herders were asked about their dietary habits in 1999 and 2002. These herders lived in areas that were severely affected by the Chernobyl accident and stated that they had reduced their intake of wild mushrooms and berries after the accident. In 2002, consumption of mushrooms in this group was approx. 1.6 kg per household per year, while the intake of berries was about 13.1 kg per household per year. Otherwise, there is little documentation of the annual intake of mushrooms and berries among Norwegians. Since the investigations of herders specified to have a lower intake of mushrooms and berries as a result of contaminated sites, it can be assumed that the consumption of other less affected areas may be higher [34].

3. Sampling

BERMUDA partners collected samples (soils, berries, mushrooms) during the summer-fall 2012. Pre-treatment is nearly completed and the samples are waiting to be analyzed. ISO standards were used at all the stages of the work if applicable (Table 9).

Table 9. Summary of the samples collected during summer-autumn 2012. NB: Some of the sampling has not yet been reported in detail and data may be missing from this table.

Sampling site	Background level	Soil	Berry	Mushroom	Other
Søve	TENORM	1	1		1
Fen gruve	TENORM	1	1		1
Rullekollen	High NORM	1	1		1
Bolledalen	High NORM	2	3	2	2
Torsnes	Normal	1	1		1
Espoo	Normal	3*	1	4	1
Kuusamo	Low-normal	3*	2		1
Loviisa	High-normal	3*	1	4	1
Bidstrup	Low	2**	2	yes	
Hallen/Oviken	High NORM	3	5	1	6
Harbo		?	?	?	?
Uppsala	Normal	?	?	?	?
Total		>20	18	>12	15

* litter, organic and mineral soil horizons separately
** organic and mineral soil horizons separately

Standardized sampling methods were used at all the stages of the work. Using ISO-standards and methods that were agreed by all partners, made sampling and pre-treatments harmonized and the work was carried out similarly in different countries. Sampled sites are approximately 100 m x 100 m (1 ha) areas and they were chosen by their suitable qualities. For example, fresh moist temperate forest with blueberry or lingonberry shrubs and a good-known mushroom area is an optimum site. All the sample species (berries and mushrooms) and soil samples were collected from this 1 ha area. Soil sampling design was random sampling of total 5 litter squares and 5 soil cores, from which organic and mineral horizons were separated. The horizons were pooled and the pooled sample was considered representative for the site.

3.1. Sites

Table 10. The sampling sites

Norway	Søve N59°16.902’ E009°17.162 Fen gruve N59°16.625’ E009°18.226’ Rullekollen N59°16.002’ E009°18.110’ Bolledalen N59°16.424 Torsnes (Reference area)
Finland	Espoo N60.335, E24.679 Kuusamo N66.292, E29.188 Loviisa N60.429, E26.318
Denmark	Bidstrup forest N 55.5558, E 11.8972
Sweden	Oviken N 63.021, E 14.393 Harbo Uppsala

In Norway two of the sites are TENORM-areas: Søve and Fen gruve. Norway also contains sites enriched in naturally occurring radionuclides (NORM), therefore Rullekollen and Bolledalen were chosen as well. The Fen area contains significant levels of NORM and the outdoor gamma doses are among the highest in Europe. Torsnes site is used as a reference area.

Two sites from southern Finland were chosen: Loviisa (N 60.429, E 26.318) where elevated radionuclide levels in soil exists and Espoo (N 60.335, E 24.679) with low levels of radionuclides. In addition, one site is located in northern Finland, in Kuusamo (N 66.292, E 29.188) where the levels are from low to normal.

The Bidstrup forest (around10 km^2) is located about 30km south of Roskilde on Zealand. The forest is close to the village of Hvalsø. It consists of a mix of hardwood and coniferous forest in a hilly terrain. The soil is clayish and rich in organic matter.

The site of Oviken is located in central Sweden. Oviken is an area with difficult varying terrain and with a high background.

3.2 Species

Together four species of berries were collected: blueberry (*Vaccinium myrtillus*), lingonberry (*Vaccinium vitis-idaea*), raspberry (*Rubus idaeus*) and blackberry (*Rubus fruticosus*). Also several species on mushrooms were collected, Chantarelle (*Cantharellus cibarius*), Funnel Chantarelle (*Cantharellus tubaeformis*), Black Chantarelle (*Craterellus cornucopioides*) and Rufous Milkcap (*Lactarius rufus*), among others.

4. Analytical methods

4.1 Pre-treatment

Berries were dried in 105 °C and then blended with mortar or a household blender for the Po/Pb-analysis. The rest of the sample was ashed for the later analysis. Mushrooms were dried about in 45 °C with a mushroom dryer and also blended with mortar or a household blender. Plant layer, litter and organic soil were air dried in <45 °C by using ISO-standard (ISO 11464:2006(E): 5.3.2 Air drying). Plants were stored for possible later use and litter was homogenized. Organic and mineral soils were 2 mm sieved and homogenized. Mineral soil was dried in 105 °C. Samples that were dried in the oven, by using standard ISO 11464:2006(E): 5.3.3 Oven drying, were spread on a tray to a layer that was not thicker than 5 cm. Additionally, archive samples for later use were stored (Table 10).

Table 10. Sample treatments and distribution of work between laboratories.

BERMUDA – Sample treatments				
Sample	**Pre-treatment**	**Amount**	**Analyser/Receiver**	**Analysis**
BERRY	1. Drying (105 °C, >24 h)	Whole sample		
	2. Blending (mortar/house hold blender)	5-30 g	STUK Rovaniemi	Po/Pb-210
	3. Ashing	1 ml	Risö/UMB. Store ash in excicator to avoid moist (hygroscopic)	ICP-MS
	4. Gamma	Rest of the sample (ashed)	STUK - PSL	Gamma
MUSHROOM	1. Drying (approx. 45 °C in mushroom dryer)	Whole sample		
	2. Blending (mortar/house hold blender)	5-30 g	STUK Rovaniemi	Po/Pb-210
	3. Ashing	1 ml	Risö/UMB. Store ash in excicator to avoid moist (hygroscopic)	ICP-MS
	4. Gamma	Rest of the sample	STUK - PSL	Gamma
SOIL				
Plant layer	1. Storing (drying 35 °C, vacuum packaging)	Whole sample	Storing for the possible later use	
Litter	1. Drying 35 °C [ISO 11464:2006(E) 5.3.2 Air drying]	Whole sample	STUK Rovaniemi	Po/Pb-210
	2. Ashing	1 ml	Risö/UMB. Store ash in excicator to avoid moist (hygroscopic)	ICP-MS
	3. Blending [ISO 11464:2006(E) (5.4.1 +)5.4.2 Crushing]	Rest of the sample	STUK - PSL	Gamma
Organic soil	1.1. pH-measurement [ISO 10390:2005(E)]	10 ml (fresh soil)		pH
	1.2. Drying 35 °C [ISO 11464:2006(E) 5.3.2 Air drying]	Whole sample	STUK Rovaniemi	Po/Pb-210
	2. Grind [ISO 11464:2006(E) (5.4.1 +) 5.4.2 Crushing]	100 ml	SLU	Soil analysis
	3. Sieving 2 mm	Rest of the sample		
	4. (Ashing possibly later)	1 ml	Risö/UMB	ICP-MS
	5. Gamma	Rest of the sample	STUK - PSL	Gamma
Mineral soil	1. Drying 35 °C [ISO 11464:2006(E) 5.3.2 Air drying]	Whole sample	STUK Rovaniemi	Po/Pb-210
	2. Grind [ISO 11464:2006(E) (5.4.1 +) 5.4.2 Crushing]	100 ml	SLU	Soil analysis
	3. Gamma (total)		STUK - PSL	Gamma
	2. Storing (dried sample, vacuum packaging)	Rest of the sample	Storing for the possible later use	
	6. (Ashing possibly later)	1 ml	Risö/UMB	ICP-MS

4.2 Measurement plan

Some analyses cannot be carried out by all partners. Therefore, samples will be sent to partners with suitable facilities. E.g ^{228}Ra and ^{226}Ra will be carried out by gamma pectroscopy at STUK, Risö and SLU, radiochemistry for ^{210}Pb and ^{210}Po will be done at STUK, UMB and Risø. Stable elements including uranium and thorium are analysed by mass spentrometry at Risø and UMB. All soil analyses (classification, soil parameters etc.) will be carried out by SLU.

The funding for the year 2013 is pending and analytical work will start as soon as funding will be confirmed.

5. Conclusions

A good set of samples has been collected and their documentation and pre-treatment has been carried out well. At this point, no results are available.

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Acknowledgements
NKS conveys its gratitude to all organizations and persons who by means of financial support or contributions in kind have made the work presented in this report possible.

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Title	Doses from natural radioactivity in wild mushrooms and berries to the Nordic population. Interim Report from the NKS-B BERMUDA activity
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ISBN	978-87-7893-346-1
Date	January 2013
Project	NKS-B / BERMUDA
No. of pages	13
No. of tables	10
No. of illustrations	1
No. of references	34
Abstract	Naturally occurring radionuclides (NORs) are the major contributors to the total effective dose of ionizing radiation of the population. Especially hazardous are the decay products of U: ^{210}Pb, ^{210}Po and ^{226}Ra in soils, water and plants. The most important exposure route to ^{210}Pb and ^{210}Po is through wild gathered food. Some studies show that ^{210}Po and also some other NORs accumulate from uranium-rich grounds in mushrooms. In berries the levels are usually lower. In Finland, Sweden and Norway there are sites enriched in NORs. In these areas e.g. the ^{210}Po levels in certain edible mushroom species may be as high as a few hundred Bq/kg, leading to effective doses of several mSv/year among certain consumer groups. The intake of wild forest products varies greatly among the population, but the public should be informed of the exposure risk and ways to minimize it. In this study, NORs and stable metals are analysed in forest soils and in common edible mushrooms and berries. Transfer factors are calculated and dose estimates from consumption of these products made. Based on the measurement data, it is estimated if highly exposed groups exist, and ways of communication with these groups will be discussed. The practical work started during summer-autumn 2012, when soils, berries and mushrooms were sampled at several sites in Finland, Norway, Denmark and Sweden, and pre-treatments carried out in the local laboratories. All sampling and pre-treatment methods were agreed by all partners and ISO standards were used when applicable, to make all stages of the work harmonized between partners. For the analytical work, samples will be sent to partners with suitable analytical facilities. The analyses will be done during the year 2013 and the results reported in the final report and in scientific publications.
Key words	Naturally occurring radionuclide, exposure, effective dose, transfer factor, berry, mushroom

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